Theory f Concentrati n P larizati n in Crossfl w Filtrati n

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A novel theory is developed for concentration polarization of non-interacting particles in crossflow-filtration systems. This theory reveals that the extent of concentration polarization, as well as the behaviour of the permeate flux, are characterized by an important dimensionless filtration number ($N_F = 4\pi a_p^3 \Delta P/3kT$). There is a critical value of the filtration number for a given suspension and operational conditions. When the filtration number is smaller than the critical value, a polarization layer exists directly over the membrane surface and the wall particle concentration is determined by the pressure and temperature. At higher filtration numbers, a cake layer of retained particles forms between the polarization layer and the membrane surface. Mathematical models are constructed for both cases and analytical solutions for the permeate flux are derived. An increase in permeate flux with increasing pressure is predicted for all operational conditions.

1. Introduction

Crossflow filtration refers to a pressure-driven separation process in which the permeate flow is perpendicular to the feed flow. Separation in crossflow filtration is usually achieved by a membrane displaying different permeability to the solvent and solute (or particles). Crossflow filtration processes are conventionally classified as reverse osmosis (hyperfiltration), ultrafiltration, and microfiltration, depending on membrane pore size and operation conditions. Crossflow filtration has marked advantages over dead-end filtration and has been widely used to separate solutes from solutions or colloidal and particulate materials from suspensions.

Concentration polarization is a phenomenon in which the solute or particle concentration in the vicinity of the membrane surface is higher than that in the bulk. This phenomenon, inherent to all crossflow-filtration processes, occurs as long as the membrane shows different permeability for the various components of the solution or suspension. The resulting concentrated layer at the membrane surface increases the filter resistance and consequently reduces the permeate flux through the membrane. Concentration polarization is of considerable interest since a high permeate rate is most desirable in filtration processes. As a result, much research effort has been expended in this area.

During the past two decades, numerous theoretical attempts have been made in an effort to understand concentration polarization and to predict permeate rate in crossflow-filtration systems. A representative achievement of these studies was development of the gel-layer model.[1,2] This model has been used as a tool for directly interpreting experimental data in ultrafiltration and as a basis for further theoretical development in microfiltration. In the latter case, shear-induced-diffusion and inertial lift are incorporated into the theory in order to obtain a better fit with experimental data.[3–5] Another popular theory in this field is the osmotic pressure model. This model, however, has been shown to be fundamentally equivalent to the gel-layer model.[6]

The gel-layer theory is derived from a single mass balance equation[1–3] with two unknowns: the permeate velocity and the particle concentration distribution over the membrane. Therefore, it is impossible to evaluate the permeate velocity from this theory without additional assumptions. The theory

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becomes a semi-empirical model as it assumes a fixed surface particle concentration and adapts a mass transfer coefficient from theories of convective heat transfer to impermeable surfaces. Neither has been theoretically proven valid for crossflow filtration. Besides fundamental shortcomings, practical applications of this model are indeed limited. Although the gel-layer model has been developed to predict the so-called 'limiting flux', the theory cannot specify explicitly the criteria necessary for achievement of limiting flux. Furthermore, the model cannot be used for conditions where the permeate velocity is pressure dependent (i.e. before limiting flux is attained).

The osmotic-pressure model has similar conceptual problems. In this model, the wall particle concentration must be estimated from certain empirical relationships or simplified assumptions. This is one of the reasons why empirical expressions are often used to predict permeate flux in reverse osmosis systems.[7,8] Evidently, this model cannot be applied to microfiltration and most ultrafiltration systems since osmotic pressure is negligible in these cases.

In this paper, a novel theory for concentration polarization is developed, based on the hydrodynamics and thermodynamics of particle suspensions. It is shown that another fundamental relationship (force or energy balance) is required, in addition to mass balance, to describe completely the concentration polarization in crossflow filtration. Similar to the gel-layer model, uniform non-interacting ('hard') spherical particles are assumed in the mathematical derivation of the theory. It is expected that this new theory will provide a solid foundation for understanding and predicting permeate flux in crossflow-filtration systems.

2. Physics in Crossflow Filtration

Crossflow-filtration systems can have different configurations (e.g. channel or tube). In the following sections, without loss of generality, a rectangular channel will be used in the theoretical analysis and a complete rejection of the solute or particles by the membrane is assumed. Several fundamental physical principles are involved in the separation process by crossflow filtration. For instance, the flow field and drag force in crossflow filtration are described by basic theories of hydrodynamics and, more importantly, many bulk properties of particle suspensions in crossflow filtration are governed by thermodynamic principles. In this section, the important features of the flow field and driving force in crossflow filtration,

3390

relevant to our theory, are briefly reviewed, followed by a discussion on the thermodynamics of crossflow-filtration systems.

2.1 Flow Field

Flow processes over the membrane surface constitute a major mechanism for concentration polarization in crossflow filtration. The transverse flow brings particles to the membrane surface and keeps the accumulated particles in the vicinity of the membrane surface. On the other hand, the longitudinal (tangent) flow sweeps accumulated particles out of the filter and tends to reduce particle concentration on the membrane surface.

The flow field in a rectangular channel with permeable wall(s) is governed, in principle, by the Navier-Stokes equation. With the assumption of constant permeation velocity along the channel, the flow field in a channel with permeable walls was first obtained by Berman.[9] It was shown that, in a thin layer near the permeable walls, the flow field at any location in the channel can be well approximated by combination of a shear tangential flow and a constant perpendicular flow.

As the suspension advances through the channel, the bulk fluid moves toward the membrane surface owing to the applied pressure. The magnitude of this transverse flow is determined by the applied pressure, the membrane resistance and the extent of concentration polarization. The permeate velocity in crossflow filtration varies along the channel length since the concentration-polarization layer thickness over the membrane surface increases gradually along the channel. However, at a given location, the flow field in a thin layer over the membrane surface can still be reasonably represented by combination of a shear longitudinal flow and a constant transverse flow.

2.2 Driving Force and Resistance

The driving force for permeate flow in crossflow filtration is the pressure drop from the bulk fluid to the permeate side of the membrane (ΔP). Particles or solutes are driven toward the membrane surface by the resulting pressure gradient. In a closed system, as the permeate passes through the membrane, the pressure gradient in the feed side of the membrane will eventually vanish because of the build-up of osmotic pressure and the filtration process will be brought to a halt. This situation occurs in dead-end filtration. Crossflow filtration represents an open system where the pressure gradient is maintained by a continuous supply of feed to the inlet and drainage of concentrated suspension from the outlet. As a result, a constant permeate flow is attained.

Permeate rate depends on the total resistance of the crossflow-filtration system. The total resistance to permeate flow is comprised of two components: (i) membrane resistance and (ii) resistance of the retained particles. Membrane resistance is usually a constant value, depending on membrane physical and chemical properties. Therefore, the flux of a pure solvent through the membrane, in the absence of retained particles, is proportional to the applied pressure. Resistance of the retained particles is attributed to the frictional drag force resulting from the passage of permeate through the dense layer of particles which are stationary in the transverse direction. This resistance is a function of the extent of particle accumulation and structure of the retained particle layer, and therefore depends on filtration performance. When the resistance is dominated primarily by the retained particles, filter performance is similar to that of an 'ideal filter' as described below.

The ideal filter displays a complete rejection of solute or particles and shows no resistance to the process liquid. In such a filter, the entire pressure drop occurs in the retained particle layer, thus greatly simplifying the theoretical analysis of concentration polarization. The concept of the ideal filter will help us to understand the fundamental mechanisms of concentration polarization in crossflow-filtration systems.

2.3 Principle of Minimum Gibbs Energy

In a crossflow-filtration system, because pressure decreases from the bulk toward the membrane surface, particles on the membrane surface will have minimum Gibbs energy. Particles will accumulate on the membrane surface because a system tends to minimize its Gibbs energy. The accumulation of particles in the vicinity of the surface is hindered by the thermal (random) motion of particles. The thermal motion of particles can be measured by the osmotic (thermodynamic) energy, which is a function of particle concentration. The gradient of the osmotic energy is from the membrane surface to the bulk fluid.

At steady state, the total energy of a retained particle on any section of the filter is equal in the transverse direction and there is no net movement of retained particles in this direction. The system is then said to reach a state of thermodynamic equilibrium. In this case, the concentration of particles on the membrane surface is uniquely determined by the pressure drop applied to the accumulated particle layer. This describes the thermodynamic foundation of concentration polarization in crossflow filtration.

Because of the finite size of particles, the particle concentration on the membrane surface reaches its maximum value (closest packing) at a certain pressure. A further increase in the applied pressure beyond this point cannot be counterbalanced by the osmotic force since it has already reached the maximum value. This situation represents a non-equilibrium (thermodynamic) system. In this case, particles in the bulk suspension have higher Gibbs energy than those on the membrane surface. The retained particles will move toward the membrane surface and form a cake layer between the polarization layer and the membrane surface. The cake layer has a porosity corresponding to the maximum packing of the retained particles. Once a cake layer forms, the pressure drop across the polarization layer will be maintained at a critical value corresponding to the porosity of a maximum packing. The excess applied pressure will be absorbed by the cake layer. This process is described as cake formation and can be considered as a special case of concentration polarization.

Since the hydrodynamics in the cake layer differ from those in the concentration polarization layer, where particle concentration is below maximum packing, theories of crossflow filtration will be developed for both cases in the following sections. A criterion for cake formation will also be given along the development of the theories.

3. Concentration Polarization

As liquid passes through the membrane, particles accumulate in the vicinity of the membrane surface and form a thin layer in which the particles are stationary in the transverse direction with respect to the membrane surface. This stationary layer of retained particles provides additional resistance to permeate flow. The resistance of the retained particle layer strongly depends on the total number of particles in this layer and on their spatial distribution. Therefore, this section will start with quantifying the number and concentration distribution of retained particles in the concentration polarization layer.

3.1 Particle Concentration Polarization Layer

Since only the accumulated particles provide additional resistance to fluid flow, we use C to denote the excess number concentration of particles in the polarization layer. The actual particle concentration is $C_0 + C$, where C_0 is the bulk (feed) number concentration of particles. The distribution of accumulated particles in the stationary layer is governed by the following equations:

$$v(x)C + D\frac{dC}{dy} = 0 \qquad (3.1)$$

$$\int_0^\infty C \, dy = M_p \qquad (3.2)$$

where x and y are the longitudinal and transverse coordinates, respectively, $v(x)$ is the permeate velocity at x, D is the particle diffusion coefficient, and M_p is the total number of excess particles accumulated over a unit membrane surface area. The direction of the permeate velocity here is defined as from the bulk to the membrane, so that $v(x)$ is always positive. The transverse coordinate y points into the flow from the membrane surface, to which we assign coordinate position $y = 0$. [In common formulations, the signs of the diffusive and convective terms in eqn. (3.1) are opposite.]

The solution of eqn. (3.1) with the mass balance condition eqn. (3.2) is

$$C = \frac{M_p v(x)}{D} \exp[-v(x)y/D] \qquad (3.3)$$

The total number of retained particles, M_p, is determined as follows.

3.2 Particle-flux Conservation

At steady state, the particle flux along the channel is a constant and is equal to the particle flux entering the inlet section of the channel. Therefore, one has the following flux conservation relationship:

$$\int_0^\infty uC \, dy + \bar{u}C_0 B = \bar{u}_0 C_0 B \qquad (3.4)$$

where u is the longitudinal velocity (here particles are assumed to take the velocity of the undisturbed flow), $\bar{u}_0$ and $\bar{u}$ are the average longitudinal velocities at the inlet section ($x = 0$) and at section of position x, respectively, and B is the channel height.

Inserting eqn. (3.3) into eqn. (3.4) and solving the resulting equation for M_p, the total number of accumulated particles (per unit area) at position x, yields

$$M_p = \frac{C_0}{D\gamma} v(x) \int_0^x v(x') \, dx' \qquad (3.5)$$

where γ is the shear rate of the longitudinal flow. Note that the shear longitudinal flow

$$u = \gamma y \qquad (3.6)$$

and the fluid mass balance

$$(\bar{u}_0 - \bar{u})B = \int_0^x v(x') \, dx' \qquad (3.7)$$

were used to reach the above solution for M_p.

The particle concentration distribution in the channel is obtained by substituting eqn. (3.5) into eqn. (3.3):

$$C = \frac{C_0}{D^2\gamma}\left[v(x)^2 \int_0^x v(x') \, dx'\right] \exp[-v(x)y/D] \qquad (3.8)$$

It seems from eqn. (3.8) that the wall particle concentration (i.e. at $y = 0$) is strongly dependent on the permeate velocity. However, as will be shown later, there is a more fundamental dependence of the wall concentration on thermodynamic variables. Even more striking is that the expression in the first square brackets of eqn. (3.8) is invariant in an ideal filter. This means that the wall concentration is not affected by the permeate velocity. We shall discuss this aspect later in more detail when the solution for the permeate velocity is available.

3.3 Crossflow-filtration Equation

As mentioned before, there are two components for the resistance to permeate flow in a crossflow-filtration system, namely the membrane resistance and the accumulated particle layer resistance. The pressure drop through the accumulated particle layer, ΔP_p, can be calculated from[10,11]

$$\Delta P_p = \int_0^{\delta_p} FC \, dy \qquad (3.9)$$

where F is the drag force exerted by a stationary retained particle on the permeate flow and δ_p is the thickness of the accumulated particle layer. The pressure drop across the membrane is simply the product of the membrane resistance and the permeate velocity. Hence, the sum of the pressure drops across the membrane and the accumulated particle layer is equal to the applied pressure. This leads to the following relationship:

$$\Delta P = \lambda v(x) + \int_0^\infty \left[\frac{kT}{D} A_s v(x)\right] C \, dy \qquad (3.10)$$

where ΔP is the pressure drop from the bulk to the permeate side of the membrane, λ is the membrane resistance, k is the Boltzmann constant, T is the absolute temperature and A_s is a correcting factor for Stokes' law. The effective applied pressure drop, ΔP, is equal to the applied hydraulic pressure minus the osmotic pressure difference between the bulk suspension and the permeate.

The expression in square brackets of eqn. (3.10) represents the drag force exerted by a stationary retained particle on the permeate flow. This expression is derived from Einstein-Stokes' law,[11] combined with Happel's cell model[12] to account for the effect of neighbouring retained particles. The term $kT/D (= 6\pi\mu a_p)$ is the friction coefficient for a spherical particle of radius a_p moving at low speed in an unbounded fluid of viscosity μ. The upper integration limit can be set to infinity because the concentration polarization layer is very thin compared to the channel height.

At high particle concentrations, as is often the case in the concentration polarization layer of crossflow filtration, the drag force exerted by a particle on the fluid deviates substantially from Stokes' law and a new relationship is required. This is usually furnished by introducing a correcting function, A_s, into Stokes' expression. Happel's cell model[12] is used to determine A_s in this study. According to the classical work of Happel[10,12]

$$A_s = \frac{1 + \frac{2}{3}\theta^5}{1 - \frac{3}{2}\theta + \frac{3}{2}\theta^5 - \theta^6} \qquad (3.11)$$

where $\theta = (1 - \varepsilon)^{1/3}$ is a porosity dependent variable, with ε being the porosity of the retained particles.

The variable θ is related to the number particle concentration:

$$\theta = (\tfrac{4}{3}\pi a_p^3 C)^{1/3} \qquad (3.12)$$

From eqn. (3.8) and (3.12), one obtains

$$v(x)C \, dy = -\frac{9D}{4\pi a_p^3} \theta^2 \, d\theta \qquad (3.13)$$

Substituting eqn. (3.13) into eqn. (3.10) and changing the corresponding integration limits results in

$$\Delta P = \lambda v(x) + \frac{kT}{D} \frac{3D}{4\pi a_p^3} \int_0^{\theta_w} A_S(\theta) 3\theta^2 \, d\theta \qquad (3.14)$$

where θ_w is the value of θ at the membrane surface. Inserting eqn. (3.8) into eqn. (3.12) and letting $y = 0$ yields

$$\theta_w = \left[\frac{4\pi C_0 a_p^3}{3D^2\gamma} v(x)^2 \int_0^x v(x') \, dx' \right]^{1/3} \qquad (3.15)$$

Eqn. (3.14) is the governing equation for the permeate velocity in crossflow filtration systems.

3.4 Solution of the Crossflow-filtration Equation

Applying the mean value theorem to eqn. (3.14) and substituting eqn. (3.15) into the resulting equation gives

$$\Delta P = \lambda v(x) + \frac{kTC_0}{D^2\gamma} A_S(\theta^*) v(x)^2 \int_0^x v(x') \, dx' \qquad (3.16)$$

where $0 < \theta^* < \theta_w$.

Let us define

$$\beta = \frac{kTC_0 A_S(\theta^*)}{D^2\gamma} \qquad (3.17)$$

Eqn. (3.16) can be then rewritten as

$$\Delta P = \lambda v(x) + \beta v(x)^2 \int_0^x v(x') \, dx' \qquad (3.18)$$

At $x = 0$, the integral in this equation vanishes, so that

$$v(0) = \frac{\Delta P}{\lambda} \qquad (3.19)$$

When $x \neq 0$, taking derivatives of both sides of eqn. (3.18) with respect to x results in

$$0 = \frac{dv(x)}{dx} \left(\lambda + 2\beta v(x) \int_0^x v(x') \, dx' \right) + \beta v(x)^3 \qquad (3.20)$$

Eliminating the integration terms from eqn. (3.18) and (3.20) yields

$$\frac{dv(x)}{dx} = -\frac{\beta v(x)^4}{2\Delta P - \lambda v(x)} \qquad (3.21)$$

Eqn. (3.21) can be integrated with eqn. (3.19) as a boundary condition and the result is

$$\left[\left(\frac{\lambda}{\Delta P} \right)^3 + \frac{6\beta}{\Delta P} x \right] v(x)^3 + 3 \frac{\lambda}{\Delta P} v(x) - 4 = 0 \qquad (3.22)$$

Eqn. (3.22) is a cubic algebraic equation of one unknown. The real root of the equation gives the general expression for the permeate velocity in crossflow-filtration systems:

$$v(x) = \frac{\Delta P}{(\lambda^3 + 6\beta \Delta P^2 x)^{1/3}}$$
$$\times \langle \{ [\lambda^3/(\lambda^3 + 6\beta \Delta P^2 x) + 4]^{1/2} + 2 \}^{1/3}$$
$$- \{ [\lambda^3/(\lambda^3 + 6\beta \Delta P^2 x) + 4]^{1/2} - 2 \}^{1/3} \rangle \qquad (3.23)$$

The average permeate flux is an important quantity which is commonly used to describe the behaviour of crossflow-

filtration systems. By definition, the average flux, V, can be calculated from

$$V = \frac{1}{L} \int_0^L v(x) \, dx \qquad (3.24)$$

where L is the channel length. A direct calculation of this integral is rather tedious because of the complex expression for $v(x)$. Fortunately, the value of this integral can be easily determined from eqn. (3.18). Thus, the average permeate flux is

$$V = \frac{\Delta P}{L\beta v(L)^2} \left[1 - \frac{\lambda}{\Delta P} v(L) \right] \qquad (3.25)$$

3.5 Filtration Number N_F and Estimation of $A_S(\theta^*)$

In order to use eqn. (3.23) and (3.25), the value of $A_S(\theta^*)$ in β has first to be determined. The estimation of $A_S(\theta^*)$ can begin from eqn. (3.14) by rewriting it as

$$\frac{4\pi a_p^3}{3kT} \Delta P_p = \int_0^{\theta_w} \frac{1 + \frac{2}{3}\theta^5}{1 - \frac{2}{3}\theta + \frac{2}{3}\theta^5 - \theta^6} 3\theta^2 \, d\theta \qquad (3.26)$$

The term on the left-hand side is a dimensionless number. We suggest to name it the filtration number for crossflow filtration and denote it as N_F. That is

$$N_F = \frac{4\pi a_p^3}{3kT} \Delta P_p \qquad (3.27)$$

The filtration number, N_F, can be considered as the ratio of the energy needed to bring a particle from the membrane surface to the bulk suspension to the thermal (dissipative) energy of the particle. A larger value of N_F indicates that more energy is needed to bring a particle from the membrane surface to the bulk suspension. As will be shown later, N_F is of fundamental importance to crossflow filtration.

For most cases where concentration polarization is appreciably developed, it is safe to neglect membrane resistance, because it is usually much smaller than the retained particle layer resistance. Thus, the value of $A_S(\theta^*)$ can be calculated by the following procedure. When operational conditions of a crossflow-filtration system are given, the filtration number, N_F, is estimated from eqn. (3.27) by substituting ΔP for ΔP_p. The value of θ_w is then determined from N_F by solving the inverse problem of eqn. (3.26). Finally, by applying the mean value theorem to eqn. (3.26) again, $A_S(\theta^*)$ is obtained from

$$A_S(\theta^*) = \frac{N_F}{\theta_w^3} \qquad (3.28)$$

An accurate value of $A_S(\theta^*)$ is expected from the above procedure as long as the resistance of the filtration system is dominated by the concentration polarization layer. When the membrane resistance is comparable with, or greater than, the resistance of the concentration polarization layer, this procedure only provides an estimate of $A_S(\theta^*)$. However, the filtration number is usually very small and $A_S(\theta^*) \approx 1$ when this situation occurs.

Values of $A_S(\theta^*)$, as well as θ_w, obtained by the procedure described above are listed in Table 1 for N_F ranging from 0 to 15.00. For a monodisperse suspension of rigid, spherical particles, the maximum packing occurs at a porosity of ca. 0.36. The corresponding N_F and θ_w values are 15.00 and 0.86. Therefore, in all situations where concentration polarization occurs, the $A_S(\theta^*)$ value can be directly obtained from Table 1. When N_F is greater than 15, the crossflow filtration becomes a non n-equilibrium system and a cake layer of retained particles will form on the membrane surface. The

Table 1 Values of θ_w and $A_s(\theta^*)$ for different filtration numbers

N_F	θ_w	$A_s(\theta^*)$
0.000	0.000	1.000
1×10^{-6}	0.002	1.002
1×10^{-5}	0.021	1.025
0.010	0.198	1.293
0.100	0.381	1.802
0.500	0.556	2.910
1.000	0.633	3.946
2.000	0.705	5.719
3.000	0.743	7.325
4.000	0.768	8.842
5.000	0.786	10.301
6.000	0.800	11.719
7.000	0.811	13.106
8.000	0.821	14.468
9.000	0.829	15.809
10.000	0.836	17.132
11.000	0.842	18.441
12.000	0.847	19.736
13.000	0.852	21.021
14.000	0.856	22.295
15.000	0.860	23.559

theory developed in this section cannot be used for such a system. A theory for non-equilibrium filtration systems will be developed in Section 4. Schematic descriptions of concentration polarization and cake formation in crossflow filtration are presented in Fig. 1.

Ranges of the filtration number for different crossflow filtration systems and typical operating conditions are listed in Table 2. It can be seen from the table that N_F for reverse osmosis is much smaller than the critical value for cake formation, while for microfiltration N_F is much greater than the
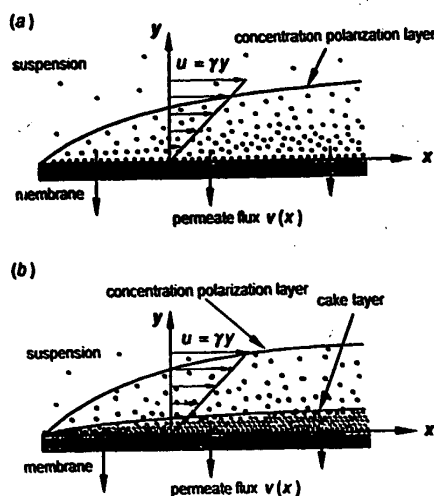

(a)

Fig. 1 Schematic description of concentration polarization and cake formation over a membrane surface in crossflow filtration. (a) Concentration polarization: there is a concentration gradient in the polarization layer. (b) Cake formation: particles accumulate in a cake layer at maximum packing.
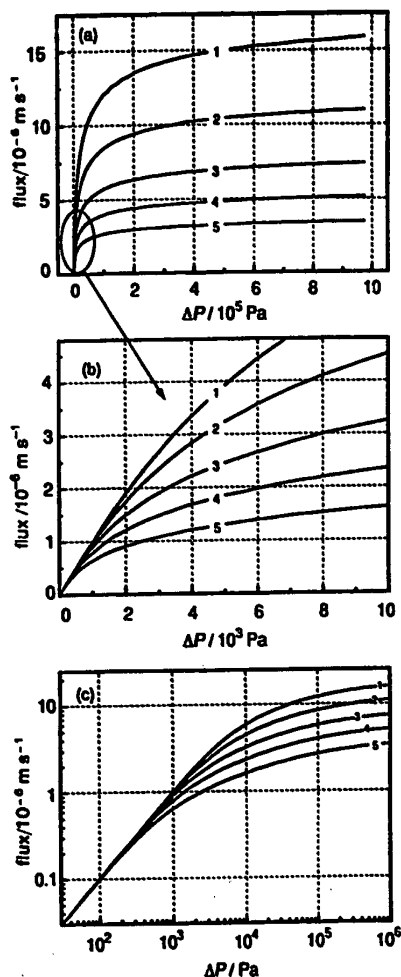

Fig. 2 Permeate flux as a function of applied pressure and bulk (feed) particle concentration. (a) Linear scale; (b) enlargement of the initial stage and (c) logarithmic scale. The curves correspond to the following bulk (feed) concentrations: 1, 1×10^{22}; 2, 3×10^{22}; 3, 1×10^{23}; 4, 3×10^{23} and 5, 1×10^{24} m^{-3}. Other parameters used in calculations: $\lambda = 109$ Pa s m^{-1}, $\gamma = 100$ s^{-1}, $a_p = 2.5 \times 10^{-9}$ m, $L = 0.5$ m and $T = 293.15$ K.

critical value. Only in some range of operating conditions of ultrafiltration, N_F is of the same order of the critical value.

3.6 Behaviour of the Permeate Flux

Average permeate fluxes as a function of pressure and feed particle concentration, calculated from eqn. (3.25), are plotted in Fig. 2 on both linear and logarithmic scales. It can be seen from Fig. 2(a), which presents a set of typical permeate–pressure curves for crossflow filtration, that there are two stages in the operation of crossflow filtration. The permeate flux changes significantly with pressure in the first stage,

Table 2 Ranges of the filtration number for different types of crossflow filtration systems

filtration system	pressure/Pa (psi)	particle size/m	N_F
reverse osmosis	10^6–4×10^6 (150–600)	3.60×10^{-10}	0.049–0.20
ultrafiltration	10^5–10^6 (15–150)	10^{-9}–10^{-7}	0.1–10^6
microfiltration	$<3 \times 10^4$ (<5)	5×10^{-8}–10^{-5}	5×10^3–5×10^{10}

while in the second stage, the flux increases much more moderately, at a rate which decreases as the pressure increases.

The dependence of permeate flux on pressure can be better observed in Fig. 2(c). There are two stages in all the curves shown in this figure. In the first stage, the flux increases roughly as the first power of the pressure. This means that concentration polarization is not significant in this range of applied pressure and the resistance to permeate flow is dominated by membrane resistance. This feature can be seen directly from eqn. (3.23). When $\lambda^3 \gg 6\beta\Delta P^2 x$ (similar to filtration of a pure solvent), we have

$$V = \frac{\Delta P}{\lambda} \qquad (3.29)$$

That is, the permeate flux is proportional to the applied pressure and to the inverse of the membrane resistance.

In the second stage, the concentration polarization is fully developed and the filter resistance is mainly determined by the resistance of the retained particle layer. This behaviour is well approximated by the ideal filter. For an ideal filter (i.e. $\lambda \to 0$), eqn. (3.23) and (3.24) can be reduced to

$$v(x) = (\tfrac{3}{2})^{1/3}(D^{2/3}\gamma^{1/3}x^{-1/3}\left(\frac{\Delta P}{A_s(\theta^*)kTC_0}\right)^{1/3} \qquad (3.30)$$

$$V = (\tfrac{3}{2})^{2/3}(D^{2/3}\gamma^{1/3}L^{-1/3}\left(\frac{\Delta P}{A_s(\theta^*)kTC_0}\right)^{1/3} \qquad (3.31)$$

The flux changes as one third power of the pressure [if the change of $A_s(\theta^*)$ with pressure is not considered]. A similar dependence of the average permeate flux on pressure in ultrafiltration systems has been recently reported.[13]

The effect of particle concentration in the feed suspension on the permeate flux can also be seen from Fig. 2. At the stage where the permeate flux is controlled by concentration polarization, the flux decreases as the feed particle concentration increases.

3.7 Concentration Distribution of Retained Particles

Combining eqn. (3.8), (3.17) and (3.18) and rearranging gives

$$C = \frac{\Delta P}{A_s(\theta^*)kT}\left[1 - \frac{\lambda}{\Delta P} v(x)\right]\exp[-v(x)y/D] \qquad (3.32)$$

Eqn. (3.32) describes the particle concentration in the entire channel. Of particular interest is the particle concentration on the membrane surface (wall), i.e., at $y = 0$. Inspection of eqn. (3.32) reveals that the wall particle concentration increases with x because the local permeate velocity $v(x)$ decreases with x. Iso-concentration curves in a crossflow filtration channel are presented in Fig. 3, on both linear and logarithmic scales. This figure indicates that the concentration polarization layer is very thin in the entire channel (of the order of 0.1 mm).

When the permeate flux is mainly controlled by the resistance of the concentration polarization layer (i.e. $\lambda^3 \ll 6\beta\Delta P^2 x$), the wall particle concentration is given by

$$C_w = \frac{\Delta P}{A_s(\theta^*)kT} \qquad (3.33)$$

Eqn. (3.33) states that when the membrane resistance can be neglected, the wall particle concentration of a given suspension is determined solely by thermodynamic variables of the system (i.e. the pressure drop ΔP and the absolute temperature T). The wall particle concentration does not change with location and, even more striking, it is independent of the bulk particle concentration. Based on eqn. (3.33), it is worth-
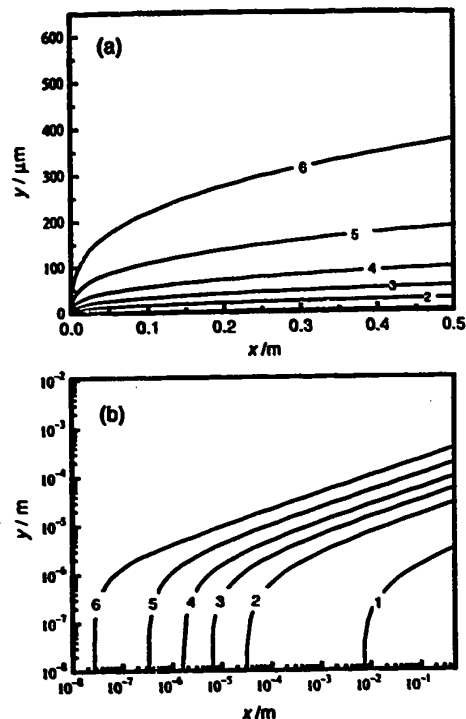

Fig. 3 Iso-concentration curves expressed as C/C_w in a rectangular channel: (a) linear scale; (b) logarithmic scale. The curves correspond to the following C/C_w values: 1, 0.95; 2, 0.70; 3, 0.50; 4, 0.30; 5, 0.10 and 6, 0.01. Parameters used in calculations: $\lambda = 10^9$ Pa s m^{-1}, $\gamma = 100$ s^{-1}, $C_0 = 10^{24}$ m^{-3}, $a_p = 2.5 \times 10^{-9}$ m, $L = 0.5$ m, and $T = 293.15$ K.

while to note that the expression in the last brackets of eqn. (3.30) and (3.31) is actually the ratio of the wall particle concentration to bulk particle concentration.

In the gel-layer model, the wall particle concentration is assumed to be a constant independent of pressure for a given type of particles. It is obvious from the above analysis that this assumption cannot be used, at least for the cases of pure concentration polarization, as otherwise it will violate the principles of thermodynamics.

4. Cake Formation

When the filtration number, N_F, is higher than ca. 15, for a monodisperse suspension of rigid spherical particles, a cake layer of retained particles will form between the concentration polarization layer and the membrane surface. By 'cake' we mean that the retained particles are packed so as to attain minimum porosity. In this section, a model for the permeate flux will be developed for cases where the additional resistance of this cake layer is considered. This theory will complement the theory developed in the previous section, and it is particularly useful for most microfiltration and some ultrafiltration processes.

4.1 Pressure Drop and Particle Distribution in the Polarization Layer

When a cake layer forms, the entire pressure drop in a crossflow-filtration system occurs at the membrane, concentration polarization layer and cake layer, i.e.

$$\Delta P = \Delta P_m + \Delta P_p + \Delta P_c \qquad (4.1)$$

where ΔP_m, ΔP_p, and ΔP_c are the pressure drops across the membrane, the polarization layer, and the cake layer, respectively. Since there is a cake layer under the polarization layer, the pressure drop through the concentration polarization layer remains at a critical value, which can be easily determined by the following equation:

$$\Delta P_p = \frac{N_{Fc} kT}{\frac{4}{3}\pi a_p^3} \qquad (4.2)$$

where N_{Fc} is the critical value of the filtration number at which the cake layer begins to appear. N_{Fc} is a function of the closest-packing porosity. When the closest-packing porosity of the retained particles is 0.36, N_{Fc} is equal to 15.00.

The distribution of retained particles in the polarization layer is similar to that without the cake layer [i.e. eqn. (3.3)], except for adjustment for the cake layer thickness in the y axis:

$$C = \frac{M_p v(x)}{D} \exp[-v(x)(y - \delta_c)/D] \qquad (4.3)$$

where δ_c and $y - \delta_c$ are the thickness of cake layer and the distance from the cake surface at x, respectively. The particle concentration in the cake layer, on the other hand, is a constant which corresponds to the maximum packing of the retained particles.

4.2 Cake Layer Thickness

Applying Stokes' law and Happel's cell model, the number of accumulated particles in the cake layer at position x, M_c, can be obtained following a procedure similar to that described in the section on concentration polarization:

$$M_c = \frac{D \Delta P_c}{kT A_s(\theta_{max}) v(x)} \qquad (4.4)$$

where θ_{max} is the θ value for maximum packing. The cake layer thickness, δ_c, can then be calculated by

$$\delta_c = \frac{\frac{4}{3}\pi a_p^3}{\theta_{max}^3} M_c = \left[\frac{\Delta P_c}{\Delta P_p}\frac{A_s(\theta_{max}^*)}{A_s(\theta_{max})}\right]\frac{D}{v(x)} \qquad (4.5)$$

The expression in the square brackets of eqn. (4.5) is another dimensionless number. This dimensionless number, denoted as N_c, is termed the cake thickness factor, i.e.

$$N_c = \frac{\Delta P_c}{\Delta P_p}\frac{A_s(\theta_{max}^*)}{A_s(\theta_{max})} \qquad (4.6)$$

The physical meaning of this number will be clear from the following derivation.

4.3 Particle-flux Conservation

Particle-flux conservation along the filter channel at steady state yields the following relationship:

$$\int_0^\infty uC \, dy = C_0 \int_0^x v(x') \, dx' \qquad (4.7)$$

The integral on the left-hand side of eqn. (4.7) represents the longitudinal flux of the retained particles in both the cake and polarization layers.

An accurate calculation of this integral is difficult at present, because very little is known about the movement of retained particles in the cake layer and the effect of the cake layer on the fl w field above it. It is assumed in this paper that the value of the left-hand side of eqn. (4.7) can be estimated through the following simplifying assumptions: (i) the velocity of the accumulated particles in the cake layer is zero (immobile cake) and (ii) the fluid field in the channel is undis-

turbed by the cake layer. These assumptions comply with the intuitive anticipation that the build-up of the cake layer will enhance the downstream movement of the retained particles.

Based on the above assumptions, the integral on the left-hand side of eqn. (4.7) can be calculated and the equation can be rewritten as

$$M_p \frac{D\gamma}{v(x)}\left[1 + \frac{v(x)\delta_c}{D}\right] = C_0 \int_0^x v(x') \, dx' \qquad (4.8)$$

Combining eqn. (4.5) and (4.8) to eliminate δ_c, and solving the resulting equation for M_p yields

$$M_p = \frac{C_0}{D\gamma(1 + N_c)} v(x) \int_0^x v(x') \, dx' \qquad (4.9)$$

It can be seen from eqn. (4.9) that the cake thickness factor, N_c, reflects the effect of the cake layer thickness on the accumulation of retained particles in the polarization layer.

4.4 Relationship between Permeate Velocity and Cake Thickness

Following a procedure similar to that used to derive eqn. (3.16), the permeate velocity can be related to the pressure drop across the polarization layer by

$$\Delta P_p = \frac{kTC_0 A_s(\theta_{max}^*)}{D^2\gamma(1 + N_c)} v(x)^2 \int_0^x v(x') \, dx' \qquad (4.10)$$

where $0 < \theta_{max}^* < \theta_{max}$. The value of $A_s(\theta_{max}^*)$ can be found in Table 1.

The solution of eqn. (4.10) gives the relationship between the local permeate velocity $v(x)$ and the cake layer thickness (represented by N_c):

$$v(x) = (\tfrac{3}{2})^{1/3}[D^{2/3}\gamma^{1/3}(1 + N_c)^{1/3}x^{-1/3}]\left(\frac{\Delta P_p}{A_s(\theta_{max}^*)kTC_0}\right)^{1/3} \qquad (4.11)$$

or

$$N_c = (\tfrac{2}{3})[D^{-2}\gamma^{-1}x]\left(\frac{\Delta P_p}{A_s(\theta_{max}^*)kTC_0}\right)^{-1} v(x)^3 - 1 \qquad (4.12)$$

Furthermore, the amount of retained particles in the cake layer can be related to the permeate velocity by combining eqn. (4.4)–(4.6) and eqn. (4.12):

$$M_c = (\tfrac{2}{3})\left(\frac{x}{D\gamma}\right)C_0 v(x)^2 - \left(\frac{\Delta P_p}{A_s(\theta_{max}^*)kT}\right)\frac{D}{v(x)} \qquad (4.13)$$

4.5 Crossflow-filtration Equation

Considering the pressure drop across the membrane and the cake layer leads to the following relationship:

$$\Delta P - \Delta P_p = \lambda v(x) + \left[\frac{kT}{D}A_s(\theta_{max})v(x)\right]M_c \qquad (4.14)$$

Substituting eqn. (4.13) into eqn. (4.14) results in

$$\Delta P^* = \lambda v(x) + \tfrac{2}{3}\beta^* x v(x)^3 \qquad (4.15)$$

where

$$\Delta P^* = \Delta P - \left[1 - \frac{A_s(\theta_{max})}{A_s(\theta_{max}^*)}\right]\Delta P_p \qquad (4.16)$$

and

$$\beta^* = \frac{kTC_0 A_s(\theta_{max})}{D^2\gamma} \qquad (4.17)$$

ΔP^* can be taken, nominally, as the total pressure drop across the membrane and the cake layer.

4.6 Solution of the Crossflow-filtration Equation

The real root of eqn. (4.15) gives the general expression for the permeate velocity in a crossflow-filtration system where a cake layer forms:

$$v(x) = \left(\frac{\Delta P^*}{3\beta^* x}\right)^{1/3}$$
$$\times \langle\{[\lambda^3/(3\beta^* \Delta P^{*2} x) + 1]^{1/2} + 1\}^{1/3}$$
$$- \{[\lambda^3/(3\beta^* \Delta P^{*2} x) + 1]^{1/2} - 1\}^{1/3}\rangle \quad (4.18)$$

If the membrane resistance is negligible (i.e. $\Delta P_c = \Delta P - \Delta P_p$), N_c can be calculated directly from eqn. (4.6). The local permeate velocity is then given by eqn. (4.11), and the average velocity is determined by:

$$V = (\tfrac{2}{3})^{2/3}[D^{2/3}\gamma^{1/3}(1 + N_c)^{1/3}L^{-1/3}]\left(\frac{\Delta P_p}{A_s(\theta_{max})kTC_0}\right)^{1/3}$$
$$(4.19)$$

As $N_c \to 0$, eqn. (4.11) and (4.19) are reduced to eqn. (3.30) and (3.31). However, when $N_c \gg 1$, (i.e. $N_F \gg N_{Fc}$), eqn. (4.11) and (4.19) become

$$v(x) = (\tfrac{2}{3})^{1/3}(D^{2/3}\gamma^{1/3}x^{-1/3})\left(\frac{\Delta P}{A_s(\theta_{max})kTC_0}\right)^{1/3} \quad (4.20)$$

and

$$V = (\tfrac{2}{3})^{2/3}(D^{2/3}\gamma^{1/3}L^{-1/3})\left(\frac{\Delta P}{A_s(\theta_{max})kTC_0}\right)^{1/3} \quad (4.21)$$

Eqn. (4.20) and (4.21) differ from eqn. (3.30) and (3.31) in that there are no pressure-dependent parameters in the former. As a result, the predicted permeate velocity from eqn. (4.13) and (4.14) will increase strictly as one third power of the pressure.

The detailed expression for the thickness of the cake layer can be obtained by combining eqn. (4.4), (4.5) and (4.18). When the membrane resistance can be neglected, the local cake layer thickness in the channel is simply given by

$$\delta_c = \left(\frac{3}{2}\right)^{1/3}\left(\frac{Dx}{\gamma}\right)^{1/3}\left(\frac{A_s(\theta_{max})kTC_0}{\Delta P_p}\right)^{1/3}\left(\frac{N_c^3}{1 + N_c}\right)^{1/3} \quad (4.22)$$

Inspection of this equation reveals that as $N_F \to N_{Fc}$, $\delta_c \to 0$. On the other hand, when $N_F \gg N_{Fc}$, δ_c increases as two thirds power of the pressure.

5. Discussion

5.1 Complete Filtration Curve

It has been known for some time that a limiting flux may exist in crossflow filtration which is independent of the applied pressure. At the same time, however, different dependences of permeate flux on pressure have also been reported.[1-5] No sound explanation for this discrepancy has been offered yet. Based on this study, it appears that the reported different dependences of permeate flux on pressure are a reflection of the different portions of complete filtration curves.

A set of complete filtration curves, calculated based on the theory presented in Sections 3 and 4, is presented in Fig. 4. There are three regions in the curves, corresponding to the principal source of filter resistance. The first is a membrane-resistance dominated region, where the flux increases proportionally to the increase in pressure. This is followed by a polarization layer-resistance dominated region, where the retained particles provide most of the resistance to permeate flow. In this region, the flux increases initially as one third power of the pressure, and the rate of increase declines in the final stage because of the significant increase of $A_s(\theta^*)$. The final region is a cake-layer-resistance dominated region, where the flux increases strictly as a cube root of the pressure.

It is shown that there is no limiting flux in crossflow filtration when the assumptions used for the cake-layer analysis hold. Flux always increases with increasing pressure, but at different rates in different regions as shown in Fig. 4. The changes in flux behaviour reflect the fact that the dominant component of the total resistance to permeate flow shifts from the membrane to the concentration polarization layer, and then to the cake layer. In the last stage of the concentration polarization resistance dominated region, the marked increase in A_s counterbalances, to a certain degree, the effect of increase in pressure. Consequently, the dependence of flux on pressure in that stage is much smaller than one third power. This is possibly the reason for introducing the concept of limiting flux in previous studies of crossflow filtration.[1-3]

5.2 Happel's Cell Model

The hydrodynamics of concentrated suspensions are quite different from those of dilute suspensions, where the drag force exerted by a stationary spherical particle on moving fluid can be accurately calculated from Stokes' law. In a concentrated particle suspension, Happel's sphere-in-cell model is reported to predict the drag force with a considerable accu-
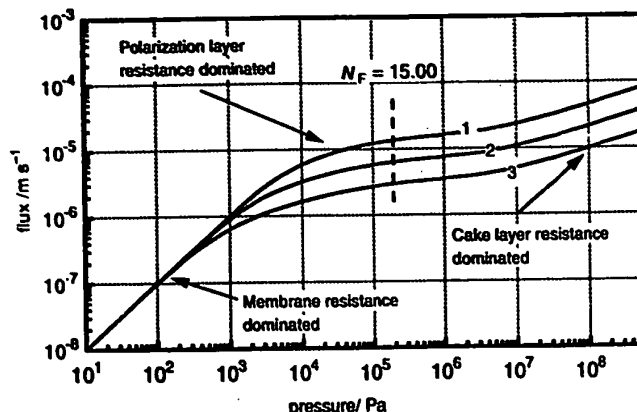

Fig. 4 'The complete filtration curve': permeate flux as a function of applied pressure for three feed (bulk) concentrations. The bulk concentrations are: 1, 1 × 10^{22}; 2, 1 × 10^{23} and 3, 1 × 10^{24} m^{-3}. The other parameters used in calculations are similar to those used in Fig. 2.

racy.[10,12] The validity of Happel's model for electrolyte solutions (as for reverse osmosis) has not, as yet, been reported in the literature.

In constructing the filtration equation [i.e. eqn. (3.10)], the key step involves relating the pressure drop across the polarization layer to the permeate velocity. This was accomplished by using Stokes' law combined with Happel's model. This analysis, however, can be carried out in a general form as follows.

Let ϕ be the chemical potential (per unit volume) of a component at concentration C. That is,

$$\phi = f(C) \tag{5.1}$$

where C is a function of the distance from the wall (membrane surface) due to concentration polarization. The osmotic (thermodynamic) force per unit volume is given by the gradient of the chemical potential:

$$F = -\frac{d\phi}{dy} \tag{5.2}$$

When the component holds a stationary position with respect to the wall surface in the transverse direction, the osmotic force must be balanced by the applied pressure. Therefore, one has

$$\Delta P - \lambda v(x) = \int_0^\infty -\frac{d\phi}{dC}\frac{dC}{dy}\,dy = \int_0^{C_w} \frac{d\phi}{dC}\,dC = \frac{d\phi}{dC}\Big|_{C=C^*} C_w$$

$$= \frac{d\phi}{dC}\Big|_{C=C^*} \frac{C_0}{D^2\gamma}\left[v(x)^2 \int_0^x v(x')\,dx'\right] \tag{5.3}$$

where C_w is the wall concentration of the component and C^* is a value between zero and C_w. In the last step of the above derivation, eqn. (3.8) is used for C_w by letting $y = 0$. Eqn. (5.3) holds for any chemical species (solute or particle) which is retained by the membrane.

Dilute suspensions of solute (or very small particles) are a special case where the osmotic energy per unit volume is given by $\phi = kTC$. Therefore, we have

$$\frac{d\phi}{dC} = kT \tag{5.4}$$

For a concentrated solution, a linear relationship does not exist between the osmotic energy and solute concentration. In this case, the value of $d\phi/dC$ can be determined by an approach similar to that used for particle suspensions, provided the relation between chemical potential and solute concentration [i.e. eqn. (5.1)] is known. For concentrated colloidal and particulate suspensions, Einstein–Stokes' law and Happel's model are adequate to relate the chemical potential to the particle concentration, which results in $d\phi/dC = A_s(\theta^*)kT$.

5.3 Units of Particle Concentration

Particle concentration is an important parameter in crossflow filtration. The volume fraction of particles is conventionally used to represent particle concentration in crossflow filtration but, as our theoretical analysis and ensuing discussion show, volume fraction is not a proper measure for particle suspensions. Most of the thermodynamic properties of suspensions, and hence the development of the concentration polarization layer in crossflow filtration, are related t the number concentration f particles. At given operating conditi ns, suspensions with similar volume fraction but different particle size can result in quite different permeate velocities, and these differences cannot be explained simply by changes in the diffusion coefficient with particle size. The theory presented in this

paper clearly shows that the resistance of the concentration polarization layer is directly related to the number concentration of retained particles through Stokes' law combined with Happel's model. Furthermore, with the number particle concentration, some important conclusions can be directly derived from thermodynamic principles. For example, when the wall concentration is below the saturation value, the number concentration, not the volume concentration, of particles is uniquely determined by the thermodynamic properties of the system. For the above reasons, the particle number concentration has been used in our theoretical development.

5.4 Wall-particle Concentration

The wall-particle concentration describes the extent of concentration polarization and can be used to predict the permeate velocity in crossflow filtration. However, based on existing concentration-polarization theories, it is not clear as yet how to determine the wall-particle concentration. In the gel-layer theory, although the wall concentration is presumed to be a constant, no theoretical methods are provided to determine the constant value. On the contrary, the osmotic pressure theory takes the wall concentration as a variable, which changes with the applied pressure. However, an expression for the wall-particle concentration in the latter case is also not available.

In this paper, we provide a clear and sound theory for calculating the wall-particle concentration. It has been shown that the wall-particle concentration is uniquely controlled by thermodynamic properties of the filtration system, characterized by the filtration number N_F. There is a critical value of the filtration number, N_{F_c}, for a given suspension. When the N_F of a crossflow-filtration system is greater than the N_{F_c} of the suspension, a cake layer of fixed particle concentration (closest packing) will form on the membrane surface. In this case, the thickness of the cake layer, rather than the concentration in the cake layer, will change with the applied pressure. On the other hand, the wall-particle concentration changes with the applied pressure when $N_F < N_{F_c}$. In this case, the wall-particle concentration is determined by the pressure drop applied to the polarization layer. If the membrane resistance to permeate flow can be neglected, the wall-particle concentration is specified by eqn. (3.33).

The possible existence of different wall-particle concentrations is a natural result of the theory presented in this paper. This theory can explain the mechanisms for the resulting wall-particle concentration in all crossflow filtration systems based on fundamental physical principles. It is, therefore, expected that this theory can be used to predict the behaviour of crossflow filtration with more confidence.

6. Summary

A comprehensive theory for concentration polarization of non-interacting particles in crossflow filtration has been presented in this paper. The essence of this theory is summarized below.

The filtration number, N_F, defined by eqn. (3.27), is one of the most important parameters in crossflow filtration. When the closest packing of a given suspension is known, there is a critical value for N_F. A cake layer will form between the concentration polarization layer and the membrane surface when N_F is greater than the critical value, while nly concentrati n polarization exists when N_F is smaller than that value. Different sets of equations are formulated to describe the processes involved in these two cases.

The filtrati n number has a simple dependence on particle size and applied pressure. For convenience, a working chart
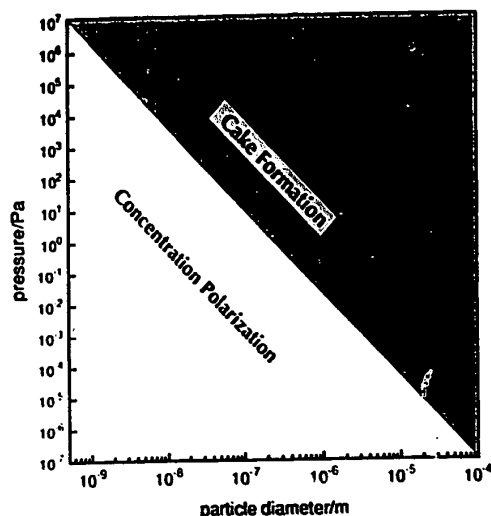
Fig. 5 A predominance diagram for concentration polarization and cake-formation regions as a function of applied pressure and particle diameter

is presented in Fig. 5, where the regions of pure concentration polarization and cake formation are displayed as a function of pressure and particle diameter. A closest packing porosity of 0.4 for uniform, spherical particles is assumed in constructing this diagram.

From this chart, one can also find the appropriate equations to use. In the pure concentration polarization region, the concentration distribution of accumulated particles in the channel is described by eqn. (3.32). The wall particle concentration is given by eqn. (3.33) when the membrane resistance is negligible. The local and average permeate fluxes are determined by eqn. (3.23) and (3.25), and approximated by eqn. (3.30) and (3.31), respectively.

In the cake-forming region, the local permeate flux is accurately given by eqn. (4.18). When the membrane resistance is negligible, the thickness of the cake layer is calculated from eqn. (4.22), and the local and average permeate fluxes are approximated by eqn. (4.11) and (4.19), respectively. When the cake layer resistance dominates, the local and average permeate fluxes can be predicted by eqn. (4.20) and (4.21), respectively.

Notation

A_s	correction function for Stokes' law based on Happel's cell model defined by eqn. (3.11)
a_p	particle radius
B	channel height
C	excess particle number concentration
C_0	bulk (feed) particle number concentration
C_w	wall particle number concentration (at $y = 0$)
D	particle diffusion coefficient; $D = kT/6\pi\mu a_p$
F	drag force exerted by a stationary particle on the permeate flow

k	Boltzmann constant, 1.3803×10^{-23} J K^{-1}
L	length of filter channel
M_c	total number of particles (per unit area) accumulated in the cake layer at position x
M_p	total number of particles (per unit area) accumulated in the polarization layer at position x
N_c	cake-forming factor defined by eqn. (4.6)
N_F	dimensionless filtration number defined by eqn. (3.27)
N_{Fc}	critical filtration number for cake forming
P	pressure
T	absolute temperature
u	axial (longitudinal) fluid velocity in the polarization layer; $u = \gamma y$
$\bar{u}$	average tangential (cross) velocity of fluid
$\bar{u}_0$	average inlet velocity of fluid (at $x = 0$)
$v(x)$	local permeate velocity (flux)
V	average permeate velocity (flux)
x	axial (longitudinal) coordinate
y	transverse coordinate
γ	fluid shear rate
β	parameter defined by eqn. (3.17)
β^*	parameter defined by eqn. (4.17)
λ	membrane resistance
μ	fluid viscosity
ε	cake or particle layer porosity
θ	parameter defined by eqn. (3.12); $\theta = (1 - \varepsilon)^{1/3}$
θ_{max}	θ value corresponding to maximum packing of the retained particles
θ_w	the value of θ at the membrane surface ($y = 0$)
θ^*	value of θ between zero and θ_w
ϕ	chemical potential (per unit volume)
δ_c	thickness of cake layer
δ_p	thickness of concentration polarization layer
ΔP	effective pressure drop (applied hydraulic pressure minus bulk osmotic pressure)
ΔP_c	pressure drop across the cake layer
ΔP_m	pressure drop across the membrane
ΔP_p	pressure drop across the concentration polarization layer
ΔP^*	a nominal pressure drop defined by eqn. (4.16)

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